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STUDY OF THE DEPENDENCE OF PHOTOENHANCED
NONLINEAR ACOUSTIC SURFACE WAVE
INTERACTIONS ON THE WAVELENGTH OF LIGHT

FINAL REPORT

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1.0 PROBLEM STUDIED

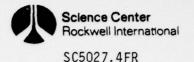
The nonlinear mixing of surface acoustic waves on a piezoelectric semiconductor is affected by the presence of band gap light. The importance of this phenomenon is that a change in optical wavelength of as little as ten angstroms has been observed to produce an output signal variation as large as 40 dB at room temperature. (1,2)

There are relatively few phenomena in nature which exhibit such narrow linewidths and large amplitude fluctuations at room temperature. During the past two years, we have been investigating the photoenhancement effect in order to 1) understand its underlying physics and 2) to extend the effect to longer optical wavelengths. This research was performed under contract numbers DAHCO4-75-C-0015 and DAAG29-75-C-0015 with the U.S. Army Research Office.

2.0 SUMMARY OF PRINCIPAL RESULTS

The principal result of our investigation up to the present is that we have shown the important role played by traps and impurity levels in the photoenhancement effect in zinc oxide. (2) We have done this by a novel application of classical transport measurements, i.e., thermally stimulated current measurements, combined with photoenhanced surface accustic wave convolution studies done as a function of varying temperature. (2)

As a result of the low temperature work, we were able to exclude the exciton resonance enhancement mechanism $^{(1)}$ as the explanation of the photoenhancement effect in zinc oxide. A different model was introduced $^{(2)}$ in which photoenhanced convolution in zinc oxide results from a nonlinear

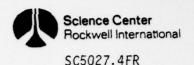


interaction between photostimulated carriers and piezoelectric surface waves coupled through the electrical conductivity of the crystal. The variation in photoenhancement with photon wavelength is then the result of corresponding variations in photoconductivity.

At this point in our work, we decided to abandon zinc oxide in favor of gallium arsenide as it is both a cleaner material and has a narrower bandgap. Convolver experiments, thermally stimulated current measurements, and photocurrent spectra were obtained on samples of semi-insulating and n^+ gallium arsenide grown at Rockwell International. The sulfur doped n^+ layer was $1.1~\mu$ thick, with a carrier concentration of about 3×10^{18} and was grown on a Cr-doped semi-insulating gallium arsenide substrate by vapor phase epitaxy. Figures 1,2, and 3 show, respectively, the thermally stimulated current for the semi-insulating material and the thermally stimulated current and photoconductivity spectrum for the n^+ epilayer material.

Surface wave convolution studies were performed on 20 mil thick (001) wafers of these materials. The input wavelength was 72 μ (38.8 MHz) and propagation was along the (110) direction. After matching the input and output, the 77.6 MHz 2 ω convolution signal was observed at the metal center plate in direct contrast with the crystal surface. However, it was found that the 2 ω convolution signal in both the Cr-doped and n⁺ epilayer GaAs samples was insensitive to bandgap light both at room temperature and at 77°K.

As can be seen from Figures 1,2, and 3, there is some photoconductivity, particularly in the n^+ epilayer material, even though no photoenhancement was seen. We explain this as follows. The open circuit rms output voltage across the convolver center plate is given by $\binom{3}{2}$

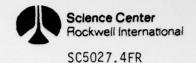


$$V_{rms} = (1/W)M(P_1P_2)^{\frac{1}{2}}$$
 (1)

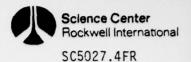
where W is the width of the center plate, P_1 and P_2 are the acoustic input powers, and M is the convolver nonlinear coupling constant $^{(3)}$ which must be either experimentally determined or theoretically calculated on the basis of a physical model. $^{(4,5)}$ In writing (1), we assume that the interacting surface waves just fill the entire area under the center plate. The coupling constant M is the sum of two terms, one due to the nonlinear piezoelectric effect which is independent of light, plus another term due to space charge nonlinearity. This term is light sensitive and is associated with the photoenhancement effect.

$$M = M \begin{pmatrix} nonlinear \\ piezoelectric coupling \end{pmatrix} + M \begin{pmatrix} space charge \\ nonlinearity \end{pmatrix}$$
 (2)

For operation at 38.8 MHz (the conditions of our experiment), the maximum value of the space charge nonlinearity term in (2) occurs for a substrate resistivity of about 1000 ohm-centimenters (see Fig. 4), $^{(4,5)}$ whereas for Cr-doped GaAs the substrate resistivity is about 10^9 ohm-cm and for the $^+$ epilayer sample it is about 10^{-3} ohm-cm. It just happens that in both cases the actual resistivity is six orders of magnitude away from the optimum value. It is clear from the space charge nonlinearity theory of convolution $^{(4,5)}$ that an order of magnitude change in resistivity for fixed acoustic input frequency corresponds to a change in M(space charge) of about an order of magnitude. Since the maximum value of M for a 38.8 MHz input frequency occurs for a 1000 ohm-cm substrate resistivity, the actual value of M attained for the Cr-doped and $^+$ GaAs substrates must have been about six orders of magnitude less than the optimum M value. For GaAs the

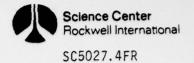


optimum value of M, $M_{\rm OPT}$ is on the order of 10^{-3} volt-meter/watt. $^{(4,5)}$ Consequently, in our experiments the space charge nonlinearity part of M must have been around 10^{-9} volt-meter/watt. Even if the effect of bandgap light increased this value of M a hundred fold, it would still only be 10^{-7} volt-meter/watt. Our experimental set up is not able to measure M values smaller than about 10^{-6} volt-meter/watt, so our failure to observe the photoenhancement effect for a 38.8 MHz input frequency on 10^{9} ohm-cm and 10^{-3} ohm-cm GaAs is understandable.



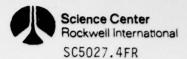
3.0 REFERENCES

- 1) T. C. Lim, T. Wolfram, E. A. Kraut, and S. K. Sinha, Wavelength dependence of the photoenhancement of nonlinear surface-wave convolution, Appl. Phys. Lett. <u>22</u>, 421 (1973).
- 2) T. C. Lim, E. A. Kraut, F. J. Morin, and J. R. Oliver, Temperature and wavelength dependence of the photoenhancement of nonlinear surface-wave convolution, Appl. Phys. Lett. 29, 229 (1976).
- 3) T. C. Lim, E. A. Kraut, and R. B. Thompson, Nonlinear materials for acoustic surface-wave convolver, Appl. Phys. Lett. <u>20</u>, 127 (1972).
- 4) O. W. Otto, Theory for nonlinear coupling between a piezoelectric surface and an adjacent semiconductor, J. of Appl. Phys. 45, 4373(1974).
- 5) H. Gautier and G. S. Kino, A detailed theory of the acoustic wave semiconductor convolver, IEEE Trans. on Sonics and Ultrasonics <u>SU-24</u>, 23 (1977).



4.0 FIGURE CAPTIONS

- Figure 1 Thermally stimulated conductivity and photoconductivity measurements on Cr-doped GaAs.
- Figure 2 Thermally stimulated conductivity and photoconductivity for n^+ epitaxial GaAs on a Cr-doped substrate.
- Figure 3 Photocurrent Spectra at 77°K for n⁺ epitaxial GaAs on a Cr-doped substrate.
- Figure 4 Convolver M value versus input frequency. Conductivity $\sigma \; \left(\text{ohm-mtr}\right)^{-1} \; \text{is a parameter.} \; \text{The curves are for a} \\ \text{degenerate GaAs convolver with the metal center plate} \\ \text{on the surface (h = 0) of the crystal.}$



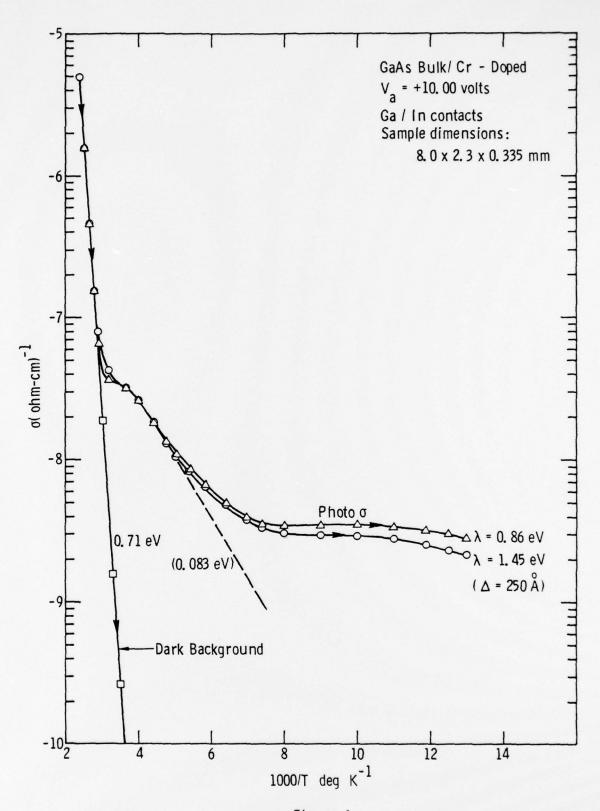
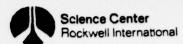


Figure 1



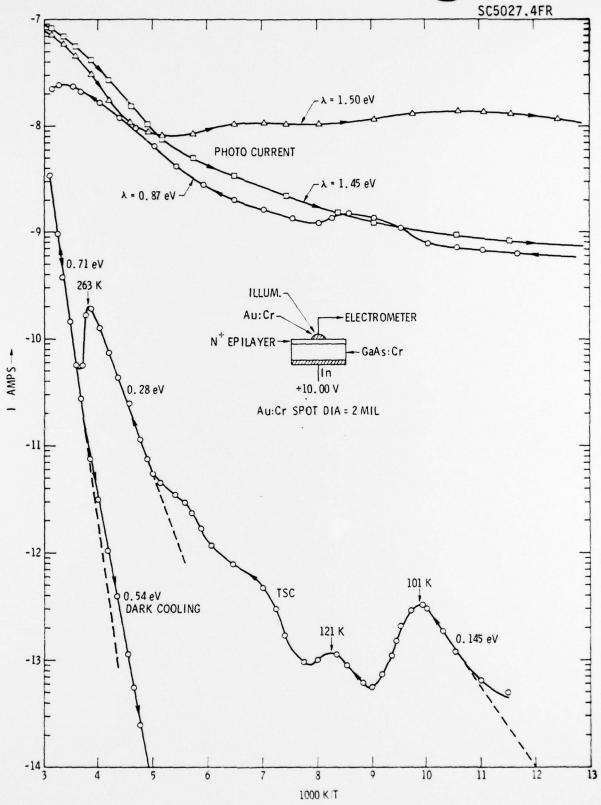


Figure 2

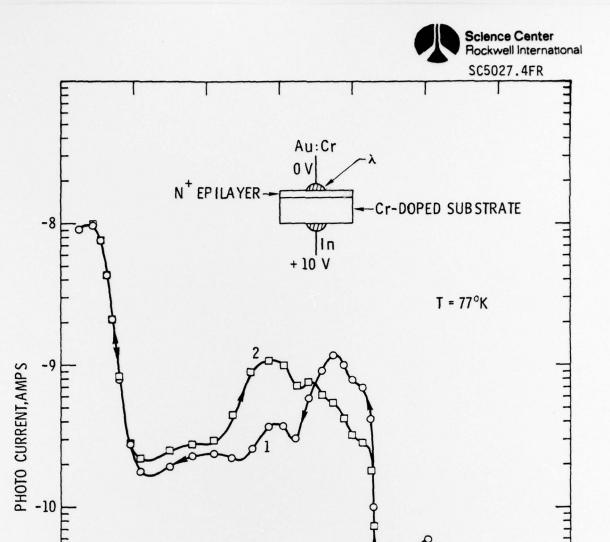


Figure 3

λ, eV

1.0

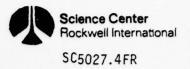
0.6

0.4

-11

-12<u>L</u>

1.4



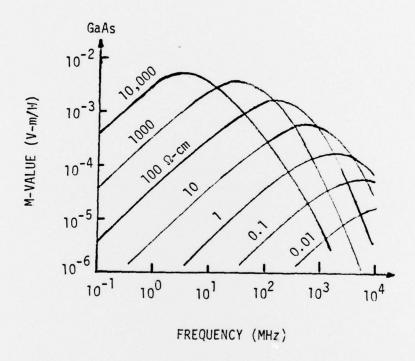


Figure 4



5.0 APPENDIX

a) Participating Scientific Personnel:

Dr. Edgar A. Kraut

Dr. T. C. Lim

Mr. Frank J. Morin

Mr. John R. Oliver

b) Publications Under the Contract:

T. C. Lim, E. A. Kraut, F. J. Morin and J. R. Oliver, "Temperature and wavelength dependence of the photoenhancement of nonlinear surface-wave convolution", Applied Physics Letters, Vol. 29, No. 4, 15 Aug. 1976, pages 229-231.

c) Travel Performed Under the Contract:

February 1976: Los Angeles to Durham, N.C.

Purpose: Technical discuss

Technical discussions with Army Research Office

personnel.

March 1977: Los Angeles to San Diego, California

Purpose: Attend American Physical Society March Meeting.

Temperature and wavelength dependence of the photoenhancement of nonlinear surface-wave convolution*

T. C. Lim, E. A. Kraut, F. J. Morin, and J. R. Oliver

Rockwell International, Science Center, P. O. Box 1085, Thousand Oaks, California 91360 (Received 3 June 1976)

Temperature-dependent structure in the photoenhancement of nonlinear surface-acoustic-wave convolution on ZnO is reported. Measurements of the temperature dependence of surface-wave convolution without light, thermally stimulated conductivity, and photoconductivity suggest that the photoenhancement effect in ZnO is sensitive to trapping and surface preparation. A possible model for the dependence of photoenhancement on photon wavelength is proposed.

PACS numbers: 78.20.Hp, 72.40.+w, 85.60.Me

Recently, we reported that photoenhancement of the convolution output of a ZnO degenerate surface-acoustic-wave (SAW) convolver exhibits maxima and minima as a function of photon wavelength. Wavelength-dependent structure in convolution output appears to be related to wavelength-dependent structure in optical absorption and reflectivity. 2-5

Similar results have been reported in $GaAs^6$ and CdS^7 . This letter presents further data on ZnO.

The configuration of the convolution device is similar to Fig. 1 of Luukkala and Kino. The lithium-doped

ZINC DXIDE PHOTOENHANCEMENT 3733A PHOTON WAVELENGTH 3690A 3996A 104°K 3692A UNITS) (RELATIVE 40004 [MENT 110.5°K EMHANCE 3700A 131°K 0.40 AAVELENGTH (MICRONS) +

FIG. 1. Photoenhanced SAW convolution output vs photon wavelength. Incident light flux decreases from 21×10^{14} photons/cm² sec at $0.42~\mu$ to 12×10^{14} photons/cm² sec at $0.36~\mu$.

ZnO crystal was supplied by Airtron, Inc. Dark resistivity exceeded $10^7~\Omega\,\mathrm{cm}$. Crystal dimensions were 3 cm long \times 8 mm wide and 1 mm thick. The normal to the 3×0.8 cm lapped surface of the crystal was within 1° of the crystal c axis. The electrical operation of the convolver and dimensions of the input transducers and center plate are similar to Ref. 1. In the present experimental arrangement the convolver is mounted on a cold finger in a liquid-nitrogen Dewar equipped with a quartz window and internal heater. The input transducers and the center plate are matched to 50 Ω . By using a monochromator (resolution of about 8 Å) to vary the wavelength of the incident light, the spectral response shown in Figs. 1 and 2 was obtained.

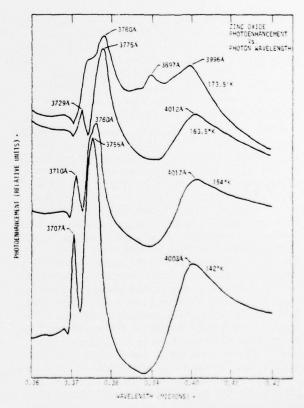


FIG. 2. Photoenhanced SAW convolution output vs photon wavelength (relative scale).

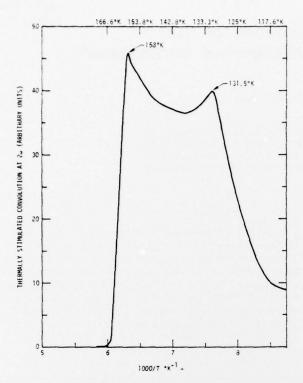


FIG. 3 Thermally stimulated SAW convolution output for ZnO in the dark vs reciprocal temperature.

The effect of temperature on convolution output without light was also determined. The convolver was
cooled to liquid-nitrogen temperatures and the traps
filled by illumination with band-gap light. The convolver
was then slowly heated in the dark. The thermally
stimulated convolution output (Fig. 3) and the photoenhanced convolution output (Figs. 1 and 2) exhibit
maximum amplitude in the same temperature range.

Photoconductivity, thermally stimulated conductivity (TSC), and decayed TSC measurements were made on lithium-doped ZnO crystals under three separate conditions; lapped surface, etched surface, and surface etched after oxidation at 700 K. Figure 4 shows the results for the case of a lapped ZnO surface with no prior treatment. The activation energies determined from the decayed TSC data are shown in Fig. 4. The photoconductivity and thermally stimulated conductivity change rapidly in the temperature range between 143 and 180°K. The same is true for the photoenhanced and thermally stimulated convolution outputs. The activation energies in Fig. 4 show that the lapped lithiumdoped ZnO sample contains many shallow traps about 0.04 to 0.05 eV below the conduction band edge. The shallow traps may be interstitial lithium donors, 10 or neutral zinc interstitials, or neutral oxygen vacancies. 11-13 In all three cases, lapped surface, etched surface, and surface etched after oxidation, there is an increase in the recombination rate above 180°K. In general, the TSC and photoconductivity data for the lapped and oxidized/etched cases show sensitivity to surface treatment. 14 A rough estimate of the trap density shows it to be the range from 10^{17} to 10^{19} depending on surface preparation.

The experimental results suggest that photoenhanced convolution in ZnO results from a nonlinear interaction between photostimulated carriers and piezoelectric surface waves coupled through the electrical conductivity of the crystal. The existence of maxima and minima in photoenhanced convolution as a function of photon wavelength may be the result of corresponding structure in the photoconductivity. An increase in the surface recombination rate relative to the bulk recombination rate is sufficient to produce a maximum in the photoconductivity. 15 For wavelengths shorter than that corresponding to the photoconductivity maximum, the photoconductivity decreases because more photons are absorbed in the surface region where the recombination rate is high and the carrier lifetime is short. Oscillatory structure in the optical absorption edge as a function of photon wavelength can give rise to additional structure in the photoconductivity. For each wavelength corresponding to an optical absorption peak, the depth of penetration of the photon into the crystal is small. Consequently, the corresponding photocarriers generated have short lifetimes and are associated with low photoconductivity. When optical absorption peaks are associated with excitons, then collisions with optical phonons or interactions with recombination centers may

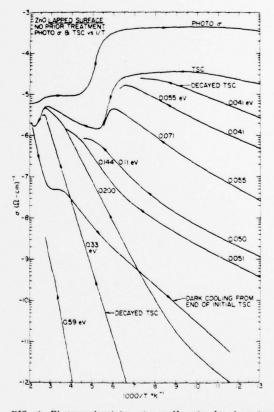


FIG. 4. Photoconductivity, thermally stimulated conductivity (TSC), and decayed TSC vs reciprocal temperature for lithium-doped ZnO (lapped surface, no prior treatment). Indicated activation energies are determined from TSC curve slopes.

cause excitons to disintegrate and contribute to the photoconductivity. 16

We have not attempted a quantitative comparison of the peaks in Figs. 1 and 2 with exciton wavelengths since the optical properties of ZnO have not been measured at the same temperatures as the photoenhancement. The observed 40-50 Å separation between the peak near 3700 Å and the peak near 3750 Å over the measured temperature range is not inconsistent with the separation between the C exciton (E||C) and the A and B excitons $(E \perp C)$. 2-5 Since at 77 °K, the A and B excitons are only about 8 Å apart, 2 it is not surprising that they are not resolved in the photoenhancement experiment. In the latter experiment the incident light was directed parallel to the C axis and therefore corresponds to the electric field being perpendicular to the Caxis. This is consistent with the amplitude ratios of the short- and intermediate-wavelength peaks in Figs. 1 and 2. The peak near 4000 Å might be associated with a bulk electron trap near 0.3 eV in Fig. 4 and reported at 0.29 eV below the conduction band edge in lithiumdoped ZnO by Seitz and Whitmore. 10

The model we propose is based on an assumed difference between the bulk and surface recombination rates for ZnO. The bulk traps are mainly shallow traps in thermal equilibrium with the conduction band and associated with multiple trapping. The surface traps correspond to a high concentration of charged lattice defects such as might be produced by surface damage.

The surface traps will be deep ones. 11 They will not be in thermal equilibrium with the conduction band and will therefore explain the assumed increase in surface recombination rate relative to the bulk.

*Work supported by the U.S. Army Research Office. ¹T.C. Lim, T. Wolfram, E.A. Kraut, and S.K. Sinha, Appl.

Phys. Lett. 22, 421 (1973).

²D.G. Thomas, J. Phys. Chem. Solids 15, 86 (1960); see also R. E. Dietz, J. J. Hopfield, and D. G. Thomas, J. Appl. Phys. Suppl. 32, 2282 (1961).

³Y. S. Park, C.W. Litton, T.C. Collins, and D.C. Reynolds, Phys. Rev. **143**, 512 (1966).

W. Y. Liang and A. D. Yoffe, Phys. Rev. Lett. 20, 59 (1968).
 T. Skettrup, Solid State Commun. 7, 869 (1969).

⁶K. Hoh, H. Hayakawa, and N. Mikoshiba, J. Appl. Phys. Supp. 2, Pt. 2, 897 (1974); J. Appl. Phys. 13, 363 (1974). 7. Ueda, J. Shirafuji, and y. Inuishi, Appl. Phys. Lett. 28, 418 (1976)

8M. Luukkala and G.S. Kino, Appl. Phys. Lett. 18, 393 (1971).

R. H. Bube, G.A. Dussel, C.T. Ho, and L.D. Miller, J. Appl. Phys. 37, 21 (1966).

¹⁰M.A. Seitz and D.H. Whitmore, J. Phys. Chem. Solids 29, 1033 (1968).

11F. A. Kroger, Chemistry of Imperfect Crystals North-Holland, Amsterdam, 1964), p. 691.

12K. I. Hagemark and L. C. Chacka, J. Solid State Chem. 15, 261 (1975).

¹³K.I. Hagemark, J. Solid State Chem. 16, 293 (1976).

1 R. J. Collins and D. G. Thomas, Phys. Rev. 112, 388 (1958).

¹⁵H.B. DeVore, Phys. Rev. 102, 86 (1956).

18E. F. Gross and B. V. Novikov, J. Phys. Chem. Solids 22, 87 (1961).

Wavelength dependence of the photoenhancement of nonlinear surface-wave convolution

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The wavelength dependence of the photoenhancement of surface-wave convolution on piezo-electric ZnO exhibits peaks which correspond to the exciton transitions observed in optical reflectivity and absorption experiments. A possible model for "exciton resonance enhancement" of nonlinear elastic-wave interactions is proposed.

In recent publications, ^{1,2} some of the present authors (T.C.L. and E.A.K.) and their co-workers estimated the magnitude of the nonlinear coupling constant of the acoustic surface-wave convolver for various piezoelectric semiconductors and piezoelectric insulators. Recently, Turner et al. ³ have observed an enhancement in the convolution output when a beam of light was shining on the center plate of a CdS degenerate convolver.

In this letter, we report on a study of the wavelength dependence of photoenhancement of convolution and second-harmonic generation on ZnO and propose a new mechanism, that of "exciton resonance enhancement", to explain the observed structure.

The configuration of the convolution device is similar to that shown in Fig. 1 of Luukkala and Kino, 4 but the ground plate is tilted 5° with respect to the center plate of the top surface so that mechanical resonance of the crystal does not occur. Two acoustic surface waves of frequency ω_s are injected at opposite ends of the crystal by means of interdigital transducers. These transducers have a wavelength of 104 µm, an aperture width of 1 mm, and 15 finger pairs and are pretuned by external inductors to give an insertion loss (ω to ω) of about 22 dB. The input electrical power to those transducers is about 100 mW. The dimensions of the center plate are 3.75 mm wide and 15 mm long, and the crystal thickness is about 1 mm. The crystal used is ZnO, and the plane of propagation is in the basal plane. For the dimensions used on the center plate, the measured capacitance (at 7 MHz) is 10 pF, and a series inductor of about 1 μH is used to tune out the capacitance of the center plate.

The unenhanced convolution output of the device is shown in Fig. 1(a). With a light source of wavelength (3771 Å) near the band-gap energy of ZnO shining vertically on the edge of the center gold plate, the convolution output is increased by about 17 dB for an input of about 7.6 $\times 10^{14}$ photons/cm² sec as shown in Fig. 1(b). The experiments were performed at room temperature ($\simeq 300$ °K).

By using a monochromator (resolution of about 8 Å) to vary the wavelength of the incident light, the spectral response shown in Fig. 2 was obtained. The output is normalized to the input optical density. $I_{2\omega}$ is the current which is due to the photoenhancement. $I_{\rm dark}$ is the current due to electroanharmonicity in the absence of light. The peak in $I_{2\omega}$ labeled C occurs at 0.377 μ m, and that labeled A, B occurs at 0.384 μ m. The peaks in $I_{2\omega}$ were observed to depend on the polarization of the light relative to the c axis (normal to the surface) of the crystal. The peak labeled C in Fig. 2 was maximum with the electric field of the light parallel to the c axis ($E \parallel c$), and minimum with polarization perpendicular to

the c axis $(E \perp c)$. We also observed a similar enhancement of the second harmonic with light incident on the surface at the gap between the transducer and center plate.

The dependence of the enhancement of the convolution output on the optical intensity shown in Fig. 3 was obtained by attenuating the incident light with a set of calibrated neutral-density filters. Additional experiments measuring current versus voltage and capacitance versus voltage across the center plate showed no effect due to the light.

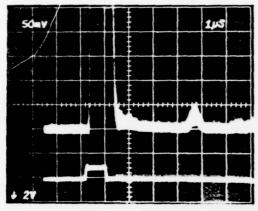
The convolution output or the nonlinear polarization voltage with no illumination depends on the contribution from the electroanharmonic constants. The mechanism of such a convolver has been explained by Kraut *et al.*² However, with near-band-gap illumination our result indicates that the nonlinear polarizability makes nonnegligible contributions due to the electro-optic as well as the acousto-optic effect.

The experimental results suggest that the peaks in the photoenhancement curve may be associated with the excitons of ZnO. We describe briefly a possible mechanism by which exciton resonance enhancement of the nonlinear electric field at acoustic frequencies can occur. It is well known that light scattering by phonons is enhanced by many orders of magnitude when the frequency of the light is near an electronic transition such as an exciton transition. The effect is called resonance Raman5,3 and Brillouin7,8 scattering for light scattered by optical and acoustic phonons, respectively. According to perturbation theory, the phenomenon results from the enhancement of the nonlinear polarizability due to intermediate electronic states (virtual transitions) which produce resonance denominators. The frequency-dependent electro-optic and elasto-optic tensors are directly related to the nonlinear polarizability and are enhanced at these optical frequencies. 6,7 In the experiment described here the enhancement of the electric field occurs at acoustic frequencies. A class of processes which produce this type of effect can be derived from the macroscopic theory of piezoelectrics.

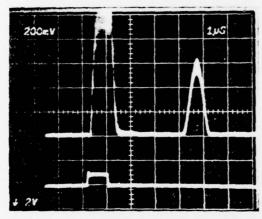
Consider two acoustic waves having frequencies ω_1 and ω_2 and wave vectors \mathbf{q}_1 and \mathbf{q}_2 and also an optical electric field \mathbf{E}_L . The nonlinear polarizability of the medium produces, by means of the resonance Brillouin effect, electric fields of the form

$$E_{\alpha}(\omega_L \pm \omega_i) = R_{\alpha\beta + \delta} S_{\beta +}^{(i)} E_{\delta}^L \exp[i(\mathbf{q}_i \pm \mathbf{q}_L) \cdot \mathbf{r} - i(\omega_i \pm \omega_L)t]$$
 (1)

(Einstein summation convention), where $S_{3\tau}^{(i)}$ are the strain components of the acoustic waves (i=1,2). The tensor R involves components of the electro-optic and



(a)



(b)

FIG. 1. Convolution output (a) without illumination and (b) with illumination of near-band-gap light. (Note the change of vertical scale.)

elasto-optic tensors, both of which are resonantly enhanced when ω_L is near an exciton transition frequency. In higher order, these two optical fields are mixed through the electro-optic effect to produce an electric field of the form

$$E_{\alpha}(\omega_{1} + \omega_{2}) = \text{Re}\left\{T_{\alpha i j k i w} S_{i j}^{(1)} S_{k i}^{(2)} E_{\lambda}^{L} E_{\nu}^{L} \exp[i(\mathbf{q}_{1} + \mathbf{q}_{2}) \cdot \mathbf{r} - i(\omega_{1} + \omega_{2}) t]\right\}. \tag{2}$$

Re indicates the real part of the bracket. The T tensor involves products of three tensor components (combination of the electro-optic and acousto-optic constants). If the acoustic waves are oppositely directed $(\mathbf{q}_1 = -\mathbf{q}_2 \text{ and } \omega_1 = \omega_2)$, then we have from Eq. (2) an electric field at $2\omega_1$ with amplitude linear in the optical intensity and proportional to the product of the strains of the acoustic waves. Very near an exciton transition the magnitude of T will be controlled by the lifetime of the exciton (which prevents the denominators from vanishing). The sketch presented here applies to bulk waves, and the extension to a careful treatment of surface waves is not trivial. However, the essential idea of exciton resonance enhancement at acoustic frequencies remains valid. There

are other mechanisms for generating nonlinear interactions. In this letter we have not attempted to describe all possible mechanisms.

According to our model, the structure (Fig. 2) observed in the wavelength dependence of the enhancement should correlate with the exciton transitions in ZnO. The exciton spectrum has been studied by Thomas9 and more recently by Park et al., 10 Liang and Yoffe, 11 and Skettrup. 12 Three strong exciton (n=1) transitions labeled A, B, and C were observed. The A and B transitions11 are excited by light having $E \perp c$ and occur at about 0.373 µm at 300 °K, while the C transition, which occurs at 0.369 μ m, is excited with $E \parallel c$. Our peaks, which occur at 0.384 and 0.377, are shifted by about 100 Å. The source of these differences is not clear at this time. The polarization effects are in agreement with the theoretical interpretation. The broad smaller maximum may be associated with the continuum of excitons above the gap or with charge carriers created by the light. The present experiment suggests that it may be possible to investigate some aspects of electronic structure by nonlinear acoustics. The sensitivity to exciton structure appears to be excellent.

Additional experiments, in particular the temperature dependence, are required to fully explore the possible role of the excitons in the photoenhancement phenomena.

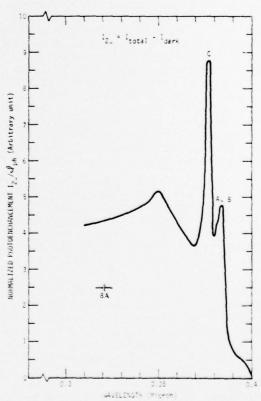


FIG. 2. Dependence of the photoenhancement effect on the wavelength of light, $I_{2\omega}$ is the current measured at the center plate and $g_{\rm ph}$ is the optical flux. Peak C corresponds to 2.16 $\mu{\rm A}$ for an input photon flux of 7.6×10^{14} photons cm² sec.

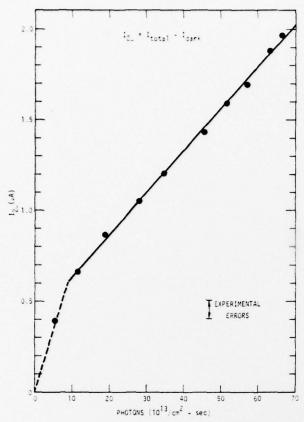


FIG. 3. Dependence of the convolution output as a function of the input photon flux. (The wavelength is 0.377 $\mu_*)$

Other II-VI compounds and alloys having strong exciton bands are expected to display similar structure. In conclusion, we would also like to point out that this acoustooptic interaction may be useful for photodetection.

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^{*}Permanent address: Ames Laboratory, U.S. Atomic Energy Commission, and Department of Physics, Iowa State University, Ames, Ia. 50010. ¹T.C. Lim, E.A. Kraut, and R.B. Thompson, Appl. Phys.

¹T.C. Lim, E.A. Kraut, and R.B. Thompson, Appl. Phys. Lett. 20, 127 (1972).

²E.A. Kraut, T.C. Lim, and B.R. Tittmann, IEEE Trans. Sonics Ultrason. 19, 247 (1972).

³C. W. Turner, I. M. Mason, and J. Chambers, Electron.

Lett. 7, 696 (1971).

⁴M. Luukkala and G.S. Kino, Appl. Phys. Lett. 18, 393 (1971).

See, for example, Proceedings of the Second International Conference on Light Scattering in Solids, Paris, 1971, edited by M. Balkanski (Flammarion Sciences, Paris, 1971), Chap.

Balkanski (Flammarion Sciences, 1971), Chap. II.

⁶R. Loudon, Proc. Roy. Soc. Lond. A 275, 218 (1963).

E. Burstein, R. Ito, A. Pinczuk, and M. Shand, J. Acoust. Soc. Am. 49, 1013 (1970).

³A.S. Pine, Phys. Rev. B 5, 3003 (1972).

³D.G. Thomas, J. Phys. Chem. Solids 15, 86 (1960); see also, R.E. Dietz, J.J. Hopfield, and D.G. Thomas, J. Appl. Phys. Suppl. 32, 2282 (1961).

¹⁰Y. S. Park, C.W. Litton, T.C. Sollins, and D.C. Reynolds, Phys. Rev. 143, 512 (1966).

W. Y. Liang and A.D. Yoffe, Phys. Rev. Lett. 20, 59 (1968).
 Skettrup, Solid State Commun. 7, 869 (1969).

APPENDIX SC5027.4FR

Nonlinear Materials for Acoustic-Surface-Wave Convolver

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Acoustic-surface-wave convolution by nonlinear mixing has been observed in cubic, hexagonal, and trigonal crystals. The magnitudes of the phenomenological nonlinear surface-wave coupling constants are tabulated and compared with the linear electromechanical coupling factor $2(\Delta V/V)$. It has been found that the basal plane of PZT-8 has a larger nonlinear coupling constant than that of the commonly used Y-cut Z-propagating LiNbO₃.

Convolution of bulk acoustic waves in piezoelectric semiconductors and piezoelectric insulators has recently been demonstrated. ^{1,2} Similar results have also been obtained ^{3,4} with elastic Rayleigh waves and recently Kraut *et al.* ⁴ have extended the work of Kirkwood ⁵ to

give a theory describing the mechanism responsible for surface-wave convolution.

Here we report experiments to evaluate the magnitude of the nonlinear coupling constants. The geometry is

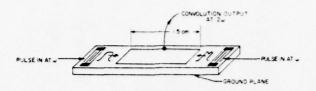


FIG. 1. Convolver configuration.

similar to the solid-center-plate configuration of Luuk-kala and Kino³ and is shown in Fig. 1. A pair of oppositely propagating acoustic surface waves of peak strain amplitude $S_1(t)$ and $S_2(t)$ and frequency ω interact in a strong piezoelectric crystal to produce an output voltage at 2ω across the center plate which, when tuned by a series inductance, is given by³

$$V_{2\omega} = \text{Re}[Av \exp(i2\omega t) \int_{t-1/2v}^{t+1/2v} S_1(T) S_2(2t-T) dT], \quad (1)$$

where4

$$A = \epsilon'_{33} \frac{W}{C} \frac{1}{H} \int_{0}^{H} {}^{1}h_{3\rho\rho km}^{s\prime} R_{\rho\rho}(x_{3}) L_{km}(x_{3}) dx_{3}, \qquad (2)$$

v is the surface-wave velocity, ϵ_{33}' is the rotated dielectric constant $(x_3$ is chosen normal to the plate), H is the crystal thickness, W is the width and l is the length of the center plate, C is the capacitance, ${}^1h_{3\rho\ell m}^{s\prime}$ is the rotated nonlinear electroanharmonic constants, 4,5 and $R_{ij}(x_3)$ and $L_{ij}(x_3)$ are the amplitudes of the strain waves traveling to the right and left, respectively. These equations assume that the width of the center plate (W) is equal to the width of the applied acoustic beam.

It is impossible at present to use Eqs. (1) and (2) to calculate voltages since a linear combination of several of the nonlinear electroanharmonic tensor components, ${}^1h_{3p'_{elbm}}^{s'}$, are involved in even the simplest cases and no unambiguous measurement of these is known to the authors. In order to evaluate materials we choose to compare the maximum received signal which occurs when pulses of equal duration are injected at each input transducer. The root-mean-square voltage $V_{\rm rms}$ at the time of maximum overlap of the input waveforms is given by

$$V_{\rm rms} = (1/W)(vt/l)M(P_1P_2)^{1/2}, \quad \text{for } t \le l/v$$
 (3)

where P_1 and P_2 are the acoustic input power during constant amplitude pulses of duration t, and M is the nonlinear coupling constant to be experimentally determined for any particular orientation. Equation (3) follows from Eqs. (1) and (2) as may be understood by noting that when the surface waves fill the area under the center plate, Eq. (3) reduces to the form $M(P_1P_2)^{1/2}/W$. The term $(1/W)(P_1P_2)^{1/2}$ determines the product of the strain amplitudes of the interacting surface waves, and M is a material constant including the electroanharmonic constants. Equation (3) applies also to cases when the sound beam does not fill the region under the center plate since the plate has been experimentally demonstrated to average the driving signal under its area. Decreasing the pulse length of constant

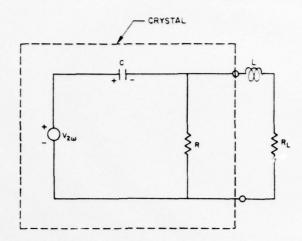


FIG. 2. Equivalent circuit of the convolver.

peak power pulses decreases the output since the constant amplitude wave occupies less area. Hence the factor vt/l appears. As the width of the input acoustic beam is decreased, keeping the input power constant, the strain amplitudes must increase within the reduced mixing area. Thus Eq. (3) is independent of the width of the input acoustic beams. It follows from Eqs. (1) and (2) that M in Eq. (3) is a material constant characteristic of the particular orientation of a particular crystal.

To determine $V_{\rm rms}$, the tuned output of the convolver, we have used a series inductance L to resonate the capacitance and observed the voltage across a $50-\Omega$ load. Measurements have been made on cubic (${\rm Bi}_{12}{\rm GeO}_{20}$), hexagonal (ZnO, PZT-8), and trigonal (quartz, LiNbO₃) crystals. The surface waves on PZT-8, LiNbO₃, ${\rm Bi}_{12}{\rm GeO}_{20}$, and ZnO were excited by 15-finger-pair $104-\mu$ -wavelength interdigital transducers of 1-mm aperture. For quartz, a 30-finger-pair $70-\mu$ -wavelength transducer of 3.3-mm aperture was used. In all cases, the center plate was 3.3 mm wide and 15 mm long. The frequency of the convolution output (2ω) ranged from 30 to 90 MHz. This frequency is low enough for the lumped equivalent circuit of Fig. 1, as shown in Fig. 2, to be reasonably applicable but high enough so that the effects

TABLE I. Magnitude of the linear and nonlinear coupling

Materials	Plane	Direction	M (10 ⁻⁴) (V m/W)	$\Delta V/V$	$\Delta V/V$ Refs.
PZT-8	Basal	All	4.95×10-4	For PZT-4 242×10-4	a
LiNbO ₃	Y cut	Z	1.21×10-4	246×10-4	b
Bi ₁₂ GeO ₂₀	(001)	(110)	1.02×10^{-4}	66×10^{-4}	c
ZnO	Basal	All	2.46×10^{-5}	44×10-4	d
Quartz	Y cut	X	< 3.3×10-6	11×10 ⁻⁴	b

^aH. Thomann, Electron. Letters 5, 25, 652 (1969).

^bW.R. Smith, H.M. Gerrad, J.H. Collins, T.M. Reeder, and H.J. Shaw, IEEE Trans. Microwave Theory Tech. MTT-17, 856 (1969).

^eE.A. Kraut, B.R. Tittmann, L.J. Graham, and T.C. Lim, Appl. Phys. Letters 17, 271 (1970).

^dJ.J. Campbell and W.J. Jones, IEEE Trans. Son. Ultrason. SU-15, 209 (1968).

of higher harmonics of the elastic plate resonances can be neglected.

The measured results are given in Table I. $2(\Delta V/V)$ in this table corresponds to the linear electromechanical coupling constant⁶ of the material. It can easily be seen that the basal plane of PZT-8 has a higher nonlinear coupling constant which is about four times larger than that of the commonly used Y-cut Z-propagating LiNbO₃. From the table it is interesting to find that the magnitude of the nonlinear coupling constant agrees well with the trend of the linear electromechanical coupling, that

is, crystals which have a strong linear coupling constant also possess a strong nonlinear coupling constant.

W.C. Wang, Appl. Phys. Letters 18, 337 (1971).

²R.B. Thompson and C.F. Quate, J. Appl. Phys. 42, 907 (1971).

³M. Luukkala and G.S. Kino, Appl. Phys. Letters 18, 393 (1971).

E.A. Kraut, T.C. Lim, and B.R. Tittmann, IEEE Trans Son. Ultrason. (to be published).

⁶C.W. Kirkwood, M.S. thesis (MIT, 1968) (unpublished). ⁶J.J. Campbell and W.J. Jones, IEEE Trans. Son. Ultrason. SU-15, 209 (1968)